Non-equilibrium quantum transport in fully interacting single-molecule nanojunctions

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Using non-equilibrium Green's functions, we derive a formula for the electron current through a lead-molecule-lead nanojunction where the interactions are not restricted to the central region, but are spread throughout the system, including the leads and the lead-molecule interfaces. The current expression consists of two sets of terms. The first set corresponds to a generalized Meir and Wingreen expression where the leads' self-energies are renormalized by the interactions crossing at the molecule-lead contacts. The second set corresponds to inelastic scattering events in the leads arising from any arbitrary interaction, including electron-electron and electron-phonon coupling, treated beyond mean-field approximations. Using different levels of approximation, we are able to recover well-known expressions for the current. We also analyse how practical calculations can be performed with our formalism by using the new concept of generalized embedding potentials.

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I. CONTEXT

Developing a theory for the non-equilibrium electronic quantum transport through nanoscale junctions is a challenging task, especially when thinking in terms of applications for nanoscale electronics. Electronic transport through nanojunctions (single-molecule junctions, for example) exhibits many important new features in comparison with conduction through macroscopic systems. This leads to promising new applications in single-molecule electronics. In particular, interactions such as Coulomb interactions between the electrons and scattering from localized atomic vibrations are critically important.

Having a simple expression for the current (or the conductance) of a nanoscale object connected to terminals is most useful. This is provided by the Landauer formula[1] which describes the current in terms of local properties (transmission coefficients) of a finite central region C and the distribution functions of the electron reservoirs connected to this region C. However, the original Landauer formulation deals only with non-interacting electrons. It has been used with success in conjunction with density-functional theory calculations for realistic nanoscale systems[2–6] since DFT maps the many-electron interacting system onto an effective single-particle problem. However there are many cases when such a single-particle approach becomes questionable[7,8]. The Landauer formula has been built upon by Meir and Wingreen[9] to extend the formalism to a central scattering region containing interactions by using the non-equilibrium Green's functions formalism. Other generalizations of Landauer-like approaches to include interactions and inelastic scattering in the region C have been developed[10–13]. However, in real systems the interaction is not confined to the central region but exists throughout the system. Accounting for the interaction along the whole system is vital[7,14,15].

In this paper, we provide a complete description which generalizes the Meir and Wingreen formalism to systems where interactions exist throughout the system, as well as at the interfaces between the central region and the electrodes. Since the choice of the location of these interfaces is purely arbitrary, and since the interactions exist everywhere, our approach is formally identical to a partition-free scheme[14,16]. While keeping the approach of the original work of Meir and Wingreen[9], we derive the most general expression of the current for the fully interacting system. From this, we recover all previously derived transport expressions or corrections when introducing the appropriate level of approximation for the interaction. Our formalism also leads to the generalization of the concept of embedding potentials when the interaction crosses at the boundaries. It therefore provides an alternative way of introducing open boundary conditions with interaction in finite-size systems.

The paper is organized as follows. In Section II, we provide the generalised current formula for fully interacting systems. We describe our model in Section II A and derive the current expression in Section II B, with full details of the calculations provided in Appendix C. The connections between more conventional results: the current at equilibrium, the current formula of Meir and Wingreen and others are given in Sections II C to II F. In section II G we describe how to apply our formalism in a specific case of interaction crossing at the contacts. Finally, we conclude and discuss extension of our work in Section III.

II. NON-EQUILIBRIUM QUANTUM TRANSPORT

A. The model

The system consists of two electrodes, left L and right R, which connect a central region C via coupling matrix elements. The interaction, which we specifically leave undefined (e.g. electron-electron or electron-phonon), is

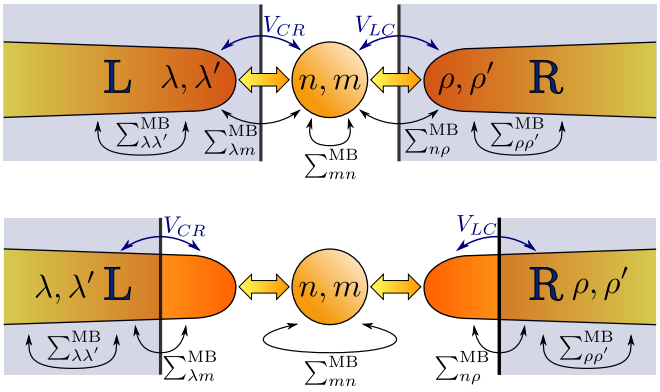

FIG. 1: Schematic representation of a central scattering region C connected to the left L and right R electrodes, with respective quantum-state labels $\{\lambda\}, \{n\}, \{\rho\}$ for the three L, C, R subspaces. Interactions are given by the coupling of the region C to the $L(R)$ electrode $V_{LC/CL}$ ($V_{RC/CR}$), and by the many-body effects $\Sigma^{\rm MB}$ within all regions as well as across the LC and CR interfaces. Top: interfaces are arbitrarily placed at the contact between the scatterer (a molecule) and the leads. They cannot be considered as being at equilibrium, being each in a region of strong spatial variation of the current and potential drop. Bottom: interfaces are now well inside the L, R regions (C region is now the so-called extended molecule), and are at local quasi-equilibrium.

spread over the entire system and crosses at the interfaces between the $L(R)$ and C regions. We use different labels for the quantum states on each side of these interfaces: $\{\lambda, \lambda'\}, \{n, m\}, \{\rho, \rho'\}$ are used to represent the complete and orthogonal set of states for the L, C and R regions respectively. We also use a compact notation for the matrix elements M of Green's functions (g, G), the self-energies (Σ) and coupling to the leads (V), where M_C represents the matrix elements M_{nm} in the region C, M_{LC} for $M_{\lambda m}$, M_{CL} for $M_{n\lambda'}$, M_{RC} for $M_{\rho m}$, M_{CR} for $M_{n\rho'}$, M_L for $M_{\lambda\lambda'}$, and M_R for $M_{\rho\rho'}$.

The complete derivation of the current expressions for the fully interacting lead-central region-lead junctions relies on only two assumptions: the many-body effects of the interacting particles are well described by self-energies $\Sigma^{\rm MB}$ in the one-particle Green's functions G, and there is no direct coupling or interaction between the states of the L and R regions: the only interaction between the leads is mediated by the region C, there is no direct coupling i.e. $\Sigma^{\rm MB}_{\lambda\rho(\rho'\lambda')} = 0$[17].

B. The non-equilibrium current

The location of the interfaces LC and CR is arbitrary (Fig. 1), and chosen purely for mathematical convenience, as the interaction spreads throughout the system. We include such interfaces to make the connection between our results and other previously derived expressions within the partitioning scheme.

From the continuity equation $\nabla\vec{j} + \partial_t n = 0$, we write the current through the interface between the L and C regions. The current flowing at the LC interface is given by $I_L(t) = -e\partial_t\langle\hat{N}_L(t)\rangle$, where $\langle\hat{N}_L(t)\rangle$ is the number of electrons in the L region, and is related to the lesser Green's function as $\langle\hat{N}_L(t)\rangle = \sum_\lambda -\mathrm{i}G^<_{\lambda\lambda}(t,t)$. From the equations of motion[18,19] obeyed by the Green's functions on the Keldysh time-loop contour C_K, we obtain the current I_L as

$$I_L(t) = \frac{e}{\hbar}\mathrm{Tr}_\lambda\left[(\Sigma G)^<(t,t) - (G\Sigma)^<(t,t)\right] \tag{1}$$

From the rules of analytical continuation on C_K (see Appendix B), we find that

$$\begin{aligned}(\Sigma G)^< &= \Sigma^{\mathrm{MB},<}G^a + (V_{LC} + \Sigma^{\mathrm{MB},r})G^<,\\ (G\Sigma)^< &= G^<(V_{CL} + \Sigma^{\mathrm{MB},a}) + G^r\Sigma^{\mathrm{MB},<}.\end{aligned} \tag{2}$$

There are no lesser (greater) components for V_{LC} since its time dependence is local $V_{LC}(t,t') = V_{LC}(t)\delta(t-t')$.

In the steady state, all double-time quantities $X(t,t')$ depend only on the time difference $X(t-t')$. The steady state current is given after Fourier transform by using $(\Sigma G)^<(t,t) \to \int \mathrm{d}\omega/2\pi\Sigma(\omega)G(\omega)$. To obtain the current, we need to calculate the following trace:

$$\begin{aligned}\mathrm{Tr}_\lambda[\dots] = &\sum_{\lambda,n,\gamma} V_{\lambda n}G^<_{n\lambda}(\omega) - G^<_{\lambda n}(\omega)V_{n\lambda}\\ &+ \Sigma^{\mathrm{MB},<}_{\lambda\gamma}(\omega)G^a_{\gamma\lambda}(\omega) + \Sigma^{\mathrm{MB},r}_{\lambda\gamma}(\omega)G^<_{\gamma\lambda}(\omega)\\ &- G^<_{\lambda\gamma}(\omega)\Sigma^{\mathrm{MB},a}_{\gamma\lambda}(\omega) - G^r_{\lambda\gamma}(\omega)\Sigma^{\mathrm{MB},<}_{\gamma\lambda}(\omega),\end{aligned} \tag{3}$$

where γ runs only on the L and C regions, since $\Sigma^{\rm MB}_{\lambda\rho} = 0$ (there is no direct coupling between the L and R regions). We then need to evaluate the Green's functions matrix elements $G^<_{n\lambda}, G^<_{\lambda n}, G^a_{n\lambda}$ and $G^r_{\lambda n}$, and $G^{<,r,a}_{\lambda\lambda'}$ by using the Dyson equation $G^x_{ij} = g^x_{ij} + [g\Sigma G]^x_{ij}$ (with $x = r, a, <$ and $\{i, j\}$ the indices for the corresponding matrix elements) and the rules of analytical continuation for the products (see Appendices B and C for detail).

We find the following general expression for the current I_L flowing through the left interface:

$$\begin{aligned}I_L = &\frac{e}{\hbar}\int\frac{d\omega}{2\pi}\\ &\mathrm{Tr}_n\left[G^r_C\tilde{\Upsilon}^l_{LC} + G^a_C(\tilde{\Upsilon}^l_{LC})^\dagger + G^<_C(\tilde{\Upsilon}_{LC} - \tilde{\Upsilon}^\dagger_{LC})\right]\\ &+\mathrm{Tr}_\lambda\left[\Sigma^{\mathrm{MB},>}_{\lambda\lambda'}G^<_{\lambda'\lambda} - \Sigma^{\mathrm{MB},<}_{\lambda\lambda'}G^>_{\lambda'\lambda}\right]\end{aligned} \tag{4}$$

where

$$\begin{aligned}\tilde{\Upsilon}_{LC} &= \Sigma^a_{CL}\,\tilde{g}^a_L\,\Sigma^r_{LC},\\ \tilde{\Upsilon}^\dagger_{LC} &= \Sigma^a_{CL}\,\tilde{g}^r_L\,\Sigma^r_{LC},\\ \tilde{\Upsilon}^l_{LC} &= \Sigma^<_{CL}\,(\tilde{g}^a_L - \tilde{g}^r_L)\,\Sigma^r_{LC} + \Sigma^r_{CL}\,\tilde{g}^<_L\,\Sigma^r_{LC}.\end{aligned} \tag{5}$$

By definition $\Sigma_{LC}(\omega) = V_{LC} + \Sigma^{\rm MB}_{LC}(\omega)$, and similarly for the CL components. $\tilde{g}^{r,a}_L(\omega)$ are the Green's functions

of the region L renormalised by the interaction *inside* that region: $(\tilde{g}_L^{r/a})^{-1} = (g_L^{r/a})^{-1} - \Sigma_L^{\mathrm{MB},r/a}$ where all quantities are defined only in the subspace L.

There are two contributions to I_L: the first trace is a generalisation of the Meir and Wingreen expression[9] to the cases where the interactions exist within the three L, C, R regions as well as in between the regions. The different quantities Υ_{LC} are related to the generalised embedding potentials (i.e. lead self-energies) with interaction crossing at the LC and CR interfaces (see end of Appendix C). The second trace in Eq. (4) is related to inelastic effects involving a sum over the states of the L region. Although the L region is semi-infinite by definition, an appropriate choice of the location of the LC interface reduces the summation. For a closed system at equilibrium, the trace $\mathrm{Tr}\left[\Sigma^{>}G^{<} - \Sigma^{<}G^{>}\right]$ is zero simply because the system obeys the detailed balance equation: $\Sigma^{>}G^{<} = \Sigma^{<}G^{>}$. For all other conditions, if the LC interface is located deep enough in the L electrode, the system is locally at quasi-equilibrium, and hence the trace vanishes (see below).

An expression similar to Eq. (4) can be obtained for the current I_R flowing at the right CR interface by swapping the index $L \leftrightarrow R$ and using the current conservation condition $I_L + I_R = 0$.

Different Green's functions

Finally we need to know, for practical calculations, the different Green's functions in all three regions. To evaluate the currents $I_{L,R}$, we need the Green's functions $G_C^{a/r,<}$ and $G_{L,R}^{a/r,<}$.

We find for $G_C^r = \langle n|G^r|m\rangle$

$$\begin{aligned} G_C^r(\omega) &= g_C^r + g_C^r\ \Sigma_C^{\mathrm{MB},r}\ G_C^r + g_C^r\ \tilde{Y}_{L+R}^r\ G_C^r \\ &= \left[(g_C^r(\omega))^{-1} - \Sigma_C^{\mathrm{MB},r}(\omega) - \tilde{Y}_{L+R}^r(\omega)\right]^{-1} \end{aligned} \tag{6}$$

where $\tilde{Y}_{L+R}^r$ is the sum of the generalised leads' self-energies $\tilde{Y}_\alpha^r$ $(\alpha = L, R)$ defined as $\tilde{Y}_\alpha^r = (\Sigma_{C\alpha}\tilde{g}_\alpha\Sigma_{\alpha C})^r$.

We find for $G_R^r = \langle\rho|G^r|\rho'\rangle$ that

$$G_{\rho\rho'}^r = \tilde{g}_{\rho\rho'}^r + \tilde{g}_{\rho\rho_1}^r\ \tilde{\tilde{Y}}_{C,\rho_1\rho_2}^r G_{\rho_2\rho'}^r \tag{7}$$

where $\tilde{\tilde{Y}}_C^r$ is the embedding potential arising from the central region C. It is defined in the right region R as follows:

$$\tilde{\tilde{Y}}_{C,\rho_1\rho_2}^r(\omega) = \Sigma_{\rho_1 m}^r(\omega)\ \tilde{\tilde{g}}_{ml}^r(\omega)\ \Sigma_{l\rho_2}^r(\omega), \tag{8}$$

with $\Sigma_{RC}^r = V_{RC} + \Sigma_{RC}^{\mathrm{MB},r}$ (similarly for Σ_{CR}^r). $\tilde{\tilde{g}}_C^r$ is a retarded Green's function of the region C renormalized by the interaction inside the central region C and by the embedding potential of the left region L only:

$$\tilde{\tilde{g}}_C^r = \left[(\tilde{g}_C^r(\omega))^{-1} - \tilde{Y}_L^r(\omega)\right]^{-1}. \tag{9}$$

The form of these equations hold for the Green's function G_L^r as well as for the advanced Green's functions $G_{L,R}^a$.

We finally get for $G_C^< = \langle n|G^<|m\rangle$:

$$G_C^< = G_C^r\ \left(\Sigma_C^{\mathrm{MB},<} + \tilde{Y}_{L+R}^<\right) G_C^a, \tag{10}$$

with $Y_{L+R}^<(\omega) = \sum_{\alpha=L,R}\left(\Sigma_{C\alpha}\tilde{g}_\alpha\Sigma_{\alpha C}\right)^<$. The rules of analytical continuation need to be applied to the products $\left(\Sigma_{C\alpha}(\omega)\tilde{g}_\alpha(\omega)\Sigma_{\alpha C}(\omega)\right)^{<,r,a}$ to get the full expansion of the generalised embedding potentials.

C. The current at equilibrium

One of the obvious checks to perform is that there is no net current at the LC and CR interfaces at equilibrium. Considering the equation for I_L given by Eq.(4), we have already shown that the trace $\mathrm{Tr}_\lambda[...]$ vanishes at equilibrium because of the detailed balance principle. Now we have to prove the same for the trace $\mathrm{Tr}_n[...]$ in Eq.(4). For this we use the procedure which consists of introducing non-equilibrium distribution functions (see Section II F below). Since at equilibrium all distributions are equal to the Fermi distribution f^{eq}, we end up, after long but trivial manipulation of Eq.(4), with

$$\begin{aligned} \mathrm{Tr}_n[...]^{\mathrm{eq}} = \mathrm{Tr}_n\big[&G_C^r\Sigma_{CL}^r\left(f^{\mathrm{eq}}\tilde{g}_L^r - \tilde{g}_L^r f^{\mathrm{eq}}\right)\Sigma_{LC}^r \\ &- G_C^a\Sigma_{CL}^a\left(f^{\mathrm{eq}}\tilde{g}_L^a - \tilde{g}_L^a f^{\mathrm{eq}}\right)\Sigma_{LC}^a\big], \end{aligned} \tag{11}$$

which after further manipulation (using complex-conjugate relations between Green's functions and self-energies) can be shown to be equal to zero. Hence, as expected, the current I_L from Eq.(4) vanishes at equilibrium.

D. Recovering the Meir and Wingreen current formula

For systems where there are interactions only within C, we have $\Sigma_{nm}^{\mathrm{MB}} \neq 0$ and $\Sigma_{LC/CL}^{a/r} = V_{LC/CL}$. Then $\tilde{g}_\alpha^x \equiv g_\alpha^x$, and $\tilde{\Upsilon}_{LC} = V_{CL}\ g_L^a\ V_{LC}$, $\tilde{\Upsilon}_{LC}^l = V_{CL}\ g_L^<\ V_{LC} = -(\tilde{\Upsilon}_{LC}^l)^\dagger$, and Eq. (4) can be recast as

$$I_L = \frac{\mathrm{i}e}{\hbar}\int\frac{\mathrm{d}\omega}{2\pi}\ \mathrm{Tr}_n\left[f_L(G_C^r - G_C^a)\Gamma_L + G_C^<\Gamma_L\right], \tag{12}$$

with $\mathrm{i}f_L\Gamma_L = V_{CL}\ g_L^<\ V_{LC}$ and $\mathrm{i}\Gamma_L = V_{CL}(g_L^a - g_L^r)V_{LC}$. Hence we recover the result of Meir and Wingreen[9].

Going one step further, we consider interaction within the L and R regions as well. The current I_L in Eq.(4) takes then the form of the Meir and Wingreen expression Eq.(12), with renormalised escape rates $\tilde{\Gamma}_L$, i.e. $\mathrm{i}\tilde{\Gamma}_L = V_{CL}\ (\tilde{g}_L^a - \tilde{g}_L^r)\ V_{LC}$ and $\tilde{\Upsilon}_{LC}^l \equiv \mathrm{i}\tilde{f}_L\tilde{\Gamma}_L(\omega) = V_{CL}\ \tilde{g}_L^<\ V_{LC}$. The interactions within the leads renormalise the coupling at the contacts $\tilde{\Gamma}_L$. Note that we have allowed for

a renormalised distribution function $\tilde{f}_L$ in the definition of $\tilde{\Upsilon}^l_{LC}$. The distribution of the left lead $\tilde{f}_L$ has the same form as the Fermi distribution function, but depending on the approximation chosen for the interaction $\Sigma^{\rm MB}_L$, the corresponding Fermi level may also need renormalization.

E. Transport with interaction on the (TD)DFT level

We consider cases where the interaction is spread throughout the entire system, and are treated at the level of density-functional theory (DFT). The exchange and correlation effects for interacting electrons are given by an effective potential $v_{xc}(r,t)$ obtained from an xc action functional of the electron density. To this potential corresponds an effective self-energy, local in both space and time[7,14]. This forms a class of self-energies, where $\Sigma^{\rm MB}(\tau,\tau') = \hat{\Sigma}^{\rm MB}(\tau)\delta(\tau-\tau')$ cannot have lesser or greater components, since the times τ and τ' must be on the same time-loop branch. With no lesser and greater components for $\Sigma^{\rm MB}$, the trace $\mathrm{Tr}_\lambda[...]$ in Eq.(4) simply vanishes. We are thus left with

$$I_L = \frac{e}{\hbar}\int\frac{\mathrm{d}\omega}{2\pi}\,\mathrm{Tr}_n\Big[G^r_C\;\tilde{\Upsilon}^l_{LC} + G^a_C\;(\tilde{\Upsilon}^l_{LC})^\dagger + G^<_C\left(\tilde{\Upsilon}_{LC} - \tilde{\Upsilon}^\dagger_{LC}\right)\Big], \quad (13)$$

where $\tilde{\Upsilon}^l_{LC} = \Sigma_{CL}\;\tilde{g}^<_L\;\Sigma_{LC}$, $(\tilde{\Upsilon}^l_{LC})^\dagger = -\Sigma_{CL}\;\tilde{g}^<_L\;\Sigma_{LC}$, $\tilde{\Upsilon}^\dagger_{LC} = \Sigma_{CL}\;\tilde{g}^r_L\;\Sigma_{LC}$, and $\Sigma = V + v_{xc}$ (V has only $V_{\alpha C/C\alpha}$ components, and v_{xc} has local static or dynamic components $v_{xc,\{\lambda,n,\rho\}}$ for DFT or time-dependent DFT calculations respectively)[20] Hence we recover a Meir-and-Wingreen-like expression for the current with renormalised $\tilde{\Gamma}_L$. The potential v_{xc} is spread throughout the system and inside the leads[7,14]. Hence Eq. (13) formally confirms the necessity of including the potential drop due to v_{xc} in the linear-response regime[21].

One should note that our formalism includes all other cases with other kind of interactions (electron-phonon) confined only in the central region[22–27]). It also includes other kind of electron-hole excitations which can be present in the leads[28]; and provides a way to treat systems with electron-hole excitations crossing at the contacts between the central region and the leads.

F. The current in terms of distribution functions and spectral densities

We now discuss when the second trace in Eq. (4) vanishes. We introduce the non-equilibrium distributions $f^<(\omega)$ obtained from the generalised Kadanoff-Baym ansatz[29] $X^<(\omega) = f^<(\omega)X^a(\omega) - X^r(\omega)f^<(\omega)$ for a Green's function or a self-energy X. We define

$$\begin{aligned}
\tilde{g}^<_L &= f^{0<}_L\tilde{g}^a_L - \tilde{g}^r_L f^{0<}_L,\\
G^<_C &= f^<_C G^a_C - G^r_C f^<_C,\\
\Sigma^<_{LC} &= f^{\rm int<}_L\Sigma^a_{LC} - \Sigma^r_{LC}f^{\rm int<}_C,\\
\Sigma^<_{CL} &= f^{\rm int<}_C\Sigma^a_{CL} - \Sigma^r_{CL}f^{\rm int<}_L
\end{aligned} \quad (14)$$

(with $f^{0<}_L$ the Fermi distribution f_L of the L region). We rewrite Eq. (4) as follows

$$\begin{aligned}
I_L = \frac{e}{\hbar}\int\frac{\mathrm{d}\omega}{2\pi}\;&\mathrm{Tr}_n\left[\delta G^<_C\;\Sigma^a_{CL}\left(\tilde{g}^a_L - \tilde{g}^r_L\right)\Sigma^r_{LC}\right]\\
&+\mathrm{Tr}_\lambda\left[\left(\Sigma^r_{LC}G^r_C\Sigma^r_{CL} - \Sigma^a_{LC}G^a_C\Sigma^a_{CL}\right)\delta\tilde{g}^<_L\right]\\
&+(2\pi)^2\mathrm{Tr}_\lambda\left[\delta f^<_L\;A^\Sigma_L(\omega)A^G_L(\omega)\right],
\end{aligned} \quad (15)$$

with $\delta g^<_L = \delta f^{0<}_L\,\tilde{g}^a_L - \tilde{g}^r_L\,\delta f^{0<}_L$, $\delta G^<_C = \delta f^<_C\,G^a_C - G^r_C\,\delta f^<_C$. The differences of distributions are $\delta f^{0<}_L = f^{0<}_L - f^{\rm int<}_L$, $\delta f^<_{L,C} = f^<_{L,C} - f^{\rm int<}_{L,C}$, and the spectral functions are $A^X_\alpha = (X^a_\alpha - X^r_\alpha)/2\pi\mathrm{i}$.

At equilibrium all distributions are equal to the Fermi distribution, all $\delta f = 0$ and $I_L = 0$ as expected. For interactions localised in C only, we again recover Eq.(12) by noticing that $\Sigma^{a/r}_{LC(CL)} = V_{LC(CL)}$, $A^\Sigma_L = 0$ and $\tilde{g}_L = g_L$. Furthermore, when the LC interface is located well inside the L region, the states λ on the left side of the interface are at their local equilibrium. Hence the corresponding distributions are equal to the local Fermi distribution and $\delta f^{0<}_L = \delta f^<_L = 0$; and thereforce the traces $\mathrm{Tr}_\lambda[...]$ in Eq. (15) and in Eq. (4) vanish (QED).

The current expression reduces then to

$$I_L = \frac{e}{\hbar}\int\frac{\mathrm{d}\omega}{2\pi}\,\mathrm{Tr}_n\left[\delta G^<_C\;\Sigma^a_{CL}\left(\tilde{g}^a_L - \tilde{g}^r_L\right)\Sigma^r_{LC}\right] \quad (16)$$

which is just another way to express the $\mathrm{Tr}_n[...]$ in Eq.(4).

G. An example of crossing interaction

We now give a brief description of how to implement our formalism for a specific case. We consider a single-molecule junction in the presence of electron-vibron interaction inside the central region and crossing at one of the contacts. We use the following Hamiltonian for the central region

$$H_C = \varepsilon_0 d^\dagger d + \omega_0 a^\dagger a + \gamma_0(a^\dagger + a)d^\dagger d \quad (17)$$

where one electronic level ε_0 and one vibration mode of energy ω_0 are coupled together via the coupling constant γ_0. The central region is coupled to the non-interacting L and R regions via hopping integrals $t_{0\alpha}$:

$$V_{LC} + V_{CR} = \sum_{\alpha=L,R} t_{0\alpha}(c^\dagger_\alpha d + d^\dagger c_\alpha). \quad (18)$$

We also consider that the hopping of an electron from the C to the L region (and *vice versa*) can excite another

vibration mode of energy ω_A via the coupling constant γ_A:

$$H_{LC} = \gamma_A(b^\dagger + b)(c_L^\dagger d + d^\dagger c_L) + \omega_A b^\dagger b. \qquad (19)$$

This model can be understood as a lowest-order expansion of the hopping integral $t_{0L}(X) = t_{0L} + t'_{0L}X$ between the C and L regions in terms of the relative position $X = \sqrt{\hbar/(2m_A\omega_A)}(b^\dagger + b)$ of the region C with respect to the region L. The Hamiltonian H_{LC} represents in this model the interaction crossing at the LC interface. The corresponding non-equilibrium Green's functions and self-energies can be calculated at different orders of the interaction using conventional non-equilibrium techniques[26,27].

Since there is no other interaction inside the L and R regions, the current expression is given by the first line of Eq.(4). We consider a mean-field approach to treat the crossing interaction. This model leads to the Hartree-like expressions for the self-energies at the LC interface:

$$\Sigma_{LC}^{\mathrm{MB},r/a} = -2\frac{\gamma_A^2}{\omega_A}\mathrm{i}\int \frac{\mathrm{d}\omega}{2\pi} G_{LC}^{<}(\omega) \qquad (20)$$

(similarly for $\Sigma_{CL}^{\mathrm{MB},r/a} \propto \int \mathrm{d}\omega G_{CL}^{<}(\omega)$).

The closed expression for $G_{LC}^{<}$ Green's function matrix elements are calculated from the corresponding Dyson equations $G_{LC}^{<} = [g\Sigma G]_{LC}^{<}$. There are no lesser and greater components for the self-energy $\Sigma_{LC}^{\mathrm{MB}}$ at the mean-field level, as we have explained in Section II E. Hence Eq.(5) reduces to

$$\begin{aligned}
\tilde{\Upsilon}_{LC} &= \Sigma_{CL}^a\, g_L^a\, \Sigma_{LC}^r, \\
\tilde{\Upsilon}_{LC}^\dagger &= \Sigma_{CL}^a\, g_L^r\, \Sigma_{LC}^r, \\
\tilde{\Upsilon}_{LC}^l &= \Sigma_{CL}^r\, g_L^<\, \Sigma_{LC}^r, \\
(\tilde{\Upsilon}_{LC}^l)^\dagger &= -\Sigma_{CL}^a\, g_L^<\, \Sigma_{LC}^a,
\end{aligned} \qquad (21)$$

with $\Sigma_{LC}^{r/a} = t_{0L} + \Sigma_{LC}^{\mathrm{MB},r/a}$.

One can see that the interaction crossing at the LC interface induces a static (however bias-dependent) renormalisation of the nominal coupling t_{0L} between the L and C regions. This non-equilibrium renormalisation induces bias-dependent modifications of the broadening of the spectral features of the C region. It can also lead to new physical non-equilibrium effects in the current[30].

The effects of the crossing interaction can also be treated beyond the mean-field level by considering a Fock-like dynamical self-energy[26,27]. In general the effects of other interaction (electron-electron) crossing at the LC and/or CL interfaces can be treated in a similar manner.

III. DISCUSSION AND CONCLUSION

We have derived a exact expression for the current through systems with interaction both within the L, C, R regions and at the LC and CR interfaces. Our result, Eq.(4), is general, assuming that there are no direct interactions between the leads; a condition that is physically sound, especially for single-molecule junctions where the spatial gap between the two electrodes is large enough. The location of the LC and CR interfaces with respect to the physical realistic scatterer is arbitrary but, in practice, should be chosen conveniently for numerical calculations. When local quasi-equilibria are reached at the interfaces, a simpler expression for the current is obtained, since the local non-equilibrium distribution functions are equal to the corresponding Fermi distributions. The deviations $\delta f^{0<}$ and $\delta f^{<}$ represent a quantitative tool to determine how far inside the leads the LC/RC interfaces need to be to reach local equilibrium. Our formalism provides a formal justification of the concept of the extended molecule that is commonly used with the conventional partitioned scheme. It also provides the correction terms needed to deal with interaction crossing at the contacts and when the contacts are not in their respective local (quasi) equilibrium.

In practice, the calculations should be performed self-consistently since the various self-energies Σ^{MB} in the three regions and at the interfaces are functionals of the all Green's functions in all the system. This offers extra degrees of freedom to perform non-fully self-consistent calculations, and test different levels of approximations for the interaction. We have given an example of how such calculations can be performed for a specific case in Section II G. We have also found that the current conservation conditions lead to an important result for a fully-interacting system: a condition that the many-body self-energies Σ^{MB} should satisfying in order to keep the conservation $I_L + I_R = 0$ (see Appendix D).

In a broader context, our formalism introduces in a formal manner the concept of generalised embedding potentials to interacting cases. Embedding methods provide the correct boundary conditions for solving the Schrödinger equation in a limited region of space, region I, automatically matching the solution on to the wavefunction in the rest of the system, region II, via the use of the embedding potential[31]. In the quantum transport community, the embedding potentials usually arise from the left and right leads to which the scatterer of interest is connected, and are commonly referred to as the lead self-energies. In conventional non-interacting approaches, they are given by $\Sigma_\alpha(\omega) = V_{C\alpha}g_\alpha(\omega)V_{\alpha C}$. In our formalism, when the interactions cross the LC/CR interfaces, we obtain a generalisation of the embedding potentials, defined as $\tilde{Y}_\alpha^x(\omega) = (\Sigma_{C\alpha}(\omega)\tilde{g}_\alpha(\omega)\Sigma_{\alpha C}(\omega))^x$. These generalised embedding potentials contain a double non-locality, in the sense that the many-body part of $\Sigma_{\alpha C}$ has a spatial extent different from that of the coupling matrix elements $V_{\alpha C}$. Hence $\tilde{Y}_\alpha$ defines a buffer zone, contained between two surfaces whose separation is related to the characteristic spatial range of the interaction self-energy $\Sigma_{\alpha C}^{\mathrm{MB}} \equiv \Sigma^{\mathrm{MB}}(|\mathbf{x}_\alpha - \mathbf{x}_n|)$. The generalised embedding potential provides a new alternative for intro-

ducing open boundary conditions with interaction within many-body finite size systems.

In summary therefore, we have introduced a new formalism for an accurate expression for the electron current in fully interacting systems. The expression is general and takes into account the fact that the interaction is crossing through the interface on which the current-density is integrated. Numerical implementations of our formalism will enable us to study cases in which the long-range Coulomb interaction is not sufficiently screened between the central region and the electrodes to be neglected or approximated, or cases in which vibration excitations at the contacts play an important role in the transport properties.

Appendix A: Relationship and symmetry on the Keldysh contour

The relations between the different components of the Green's functions and self-energies on the Keldysh time-loop contour C_K are given by:

$$\begin{aligned} X^r &= X^{++} - X^{+-} = X^{-+} - X^{--} \\ X^a &= X^{++} - X^{-+} = X^{+-} - X^{--} \\ X^{++} + X^{--} &= X^{+-} + X^{-+} \\ X^{-+} - X^{+-} &= X^r - X^a, \end{aligned} \tag{A1}$$

with $X^{\eta_1\eta_2}(12) \equiv G^{\eta_1\eta_2}(12)$ or $\Sigma^{\eta_1\eta_2}(12)$, and where $(i = 1, 2)$ is the composite index for space-time location $(\mathbf{x}_i, t_i)$ and η_i is the index of the Keldysh time-loop contour C_K branch (+ forward time arrow, − backward time arrow) on which the time t_i is located. The usual lesser and greater projections are defined respectively as $X^< \equiv X^{+-}$ and $X^> \equiv X^{-+}$, and the usual time-ordered (anti-time-ordered) as $X^t = X^{++}$ ($X^{\bar{t}} = X^{--}$).

By definition, complex conjugation of the different Green's functions follows the rules:

$$\begin{aligned} G^a(1,2) &= (G^r(2,1))^* \\ G^{\gtrless}(1,2) &= -\left(G^{\gtrless}(2,1)\right)^* \end{aligned}$$

Similar expressions hold for the self-energies $\Sigma^x(1,2)$.

Appendix B: Rules for analytical continuation

The rules for analytical continuation from C_K to normal real-time make that the following products $P_{(i)}(\tau, \tau')$ on the time-loop contour,

$$\begin{aligned} P_{(2)} &= \int_{C_K} AB \\ P_{(3)} &= \int_{C_K} ABC \\ P_{(n)} &= \int_{C_K} A_1 A_2 ... A_n, \end{aligned} \tag{B1}$$

have the following components $P^x_{(i)}(t, t')$ on the real-time axis $(x = r, a, >, <)$

$$\begin{aligned} P^{\gtrless}_{(2)} &= \int_t A^r B^{\gtrless} + A^{\gtrless} B^a \\ P^{\gtrless}_{(3)} &= \int_t A^{\gtrless} B^a C^a + A^r B^{\gtrless} C^a + A^r B^r C^{\gtrless} \\ P^r_{(n)} &= \int_t A^r_1 A^r_2 ... A^r_n \qquad P^a_{(n)} = \int_t A^a_1 A^a_2 ... A^a_n. \end{aligned} \tag{B2}$$

Appendix C: Derivation of the current I_L

In this appendix, we provide the details of the derivation of the main results of this paper, mainly Eq. (4) and Eq. (5). In the following, we choose to use the symbol $\sum\!\!\!\!\!\!\bullet$ for summations in order to have a better graphical distinction between the sum-signs and the self-energies Σ.

From Eq. (1) and Eq. (3), we need to calculate the following traces:

$$\begin{aligned} &\mathrm{Tr}_\lambda\left[(\Sigma G)^<\right] = \\ &\quad \sum_{\lambda,n}\left((V_{\lambda n} + \Sigma^{\mathrm{MB}}_{\lambda n})G_{n\lambda}\right)^< + \sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}(\Sigma^{\mathrm{MB}}_{\lambda\lambda'}G_{\lambda'\lambda})^<, \end{aligned} \tag{C1}$$

and similarly

$$\begin{aligned} &\mathrm{Tr}_\lambda\left[(G\Sigma)^<\right] = \\ &\quad \sum_{\lambda,n}\left(G_{\lambda n}(\Sigma^{\mathrm{MB}}_{n\lambda} + V_{n\lambda})\right)^< + \sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}(G_{\lambda\lambda'}\Sigma^{\mathrm{MB}}_{\lambda'\lambda})^< \end{aligned} \tag{C2}$$

since $\Sigma^{\mathrm{MB}}_{\lambda\rho} = 0$ and $\Sigma^{\mathrm{MB}}_{\rho\lambda} = 0$.

We first consider the sums $\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}$

$$\begin{aligned} &\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}(\Sigma^{\mathrm{MB}}_{\lambda\lambda'}G_{\lambda'\lambda})^< - \sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}(G_{\lambda\lambda'}\Sigma^{\mathrm{MB}}_{\lambda'\lambda})^< = \\ &\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}\Sigma^{\mathrm{MB},<}_{\lambda\lambda'}G^a_{\lambda'\lambda} + \Sigma^{\mathrm{MB},r}_{\lambda\lambda'}G^<_{\lambda'\lambda} - G^<_{\lambda\lambda'}\Sigma^{\mathrm{MB},a}_{\lambda'\lambda} - G^r_{\lambda\lambda'}\Sigma^{\mathrm{MB},<}_{\lambda'\lambda} \\ =&\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}\Sigma^{\mathrm{MB},<}_{\lambda\lambda'}(G^a_{\lambda'\lambda} - G^r_{\lambda'\lambda}) + (\Sigma^{\mathrm{MB},r}_{\lambda\lambda'} - \Sigma^{\mathrm{MB},a}_{\lambda'\lambda})G^<_{\lambda'\lambda} \\ =&\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}\Sigma^{\mathrm{MB},<}_{\lambda\lambda'}(G^< - G^>)_{\lambda'\lambda} + (\Sigma^{\mathrm{MB},>} - \Sigma^{\mathrm{MB},<})_{\lambda\lambda'}G^<_{\lambda'\lambda} \\ =&\sum\!\!\!\!\!\!\bullet_{\lambda,\lambda'}\Sigma^{\mathrm{MB},>}_{\lambda\lambda'}G^<_{\lambda'\lambda} - \Sigma^{\mathrm{MB},<}_{\lambda\lambda'}G^>_{\lambda'\lambda} \\ =&\mathrm{Tr}_\lambda\left[\Sigma^{\mathrm{MB},>}_L G^<_L - \Sigma^{\mathrm{MB},<}_L G^>_L\right]. \end{aligned} \tag{C3}$$

In the first line of Eq. (C3), we used the rules of analytical continuation. In the second, we have used the equivalent of cyclic permutation in the calculation of a trace, i.e. swapping the index λ and λ' in the last two terms. This is possible here since the sums and all matrix

elements are defined in the single subspace of the L electrode. The final result looks like the collision terms usually obtained in the derivation of a generalised Boltzmann equation from quantum kinetic theory. They correspond to the particle production (scattering-in) and absorption or hole production (scattering-out) related to inelastic processes (i.e. non-diagonal elements of the self-energy on the time-loop contour $\Sigma^<$) occuring in the left electrode.

Now we consider the sums $\sum_{\lambda,n}$ in Eq.(C1) and Eq.(C2). We find that

$$\sum_{\lambda n}[...] = \sum_{\lambda n}\left[\Sigma^r_{\lambda n}G^<_{n\lambda} - G^<_{\lambda n}\Sigma^a_{n\lambda} + \Sigma^{\mathrm{MB}<}_{\lambda n}G^a_{n\lambda} - G^r_{\lambda n}\Sigma^{\mathrm{MB}<}_{n\lambda}\right]. \tag{C4}$$

We now need to calculate the following different Green's functions matrix elements $G^<_{n\lambda}, G^<_{\lambda n}, G^a_{n\lambda}$ and $G^r_{\lambda n}$. For this we use the Dyson-like equation defined for the non-diagonal elements: $G^x_{n\lambda} = \langle n|(G\Sigma g)^x|\lambda\rangle$, and $G^x_{\lambda n} = \langle\lambda|(G\Sigma g)^x|n\rangle$, and

We concentrate on one matrix element $\langle n|(G\Sigma g)^<|\lambda\rangle$ to show the mechanism of the derivation:

$$\begin{aligned} G^<_{n\lambda} &= \langle n|(G\Sigma g)^<|\lambda\rangle \\ &= \langle n|G^r\Sigma^< g^r + G^<\Sigma^a g^a + G^r\Sigma^r g^<|\lambda\rangle \\ &= \sum_{\lambda_1,\lambda_2,m} G^r_{n\lambda_1}\,\Sigma^<_{\lambda_1\lambda_2}\,g^a_{\lambda_2\lambda} + G^r_{nm}\,\Sigma^<_{m\lambda_2}\,g^a_{\lambda_2\lambda} \\ &\quad + G^<_{n\lambda_1}\,\Sigma^a_{\lambda_1\lambda_2}\,g^a_{\lambda_2\lambda} + G^<_{nm}\,\Sigma^a_{m\lambda_2}\,g^a_{\lambda_2\lambda} \\ &\quad + G^r_{n\lambda_1}\,\Sigma^r_{\lambda_1\lambda_2}\,g^<_{\lambda_2\lambda} + G^r_{nm}\,\Sigma^r_{m\lambda_2}\,g^<_{\lambda_2\lambda}, \end{aligned} \tag{C5}$$

with $\Sigma^{a/r}_{m\lambda} = V_{m\lambda} + \Sigma^{\mathrm{MB},a/r}_{m\lambda}$ and $\Sigma^<_{m\lambda} = \Sigma^{\mathrm{MB},<}_{m\lambda}$, and, as explained above, we have used the condition $\Sigma^x_{\rho\lambda} = 0$. The same principle holds for the derivation of the other Green's functions matrix elements.

The interesting point is that the terms in $\Sigma^{a/r}_{\lambda_1\lambda_2}$ can be factorised out and included within the renormalisation of the left lead Green's functions $g^{a/r,<}_{\lambda_1\lambda_2}$ as follows

$$g^a_{\lambda\lambda'}\left(1 - \Sigma^{\mathrm{MB},a}g^a\right)^{-1}_{\lambda'\lambda_1} = \tilde{g}^a_{\lambda\lambda_1}. \tag{C6}$$

Therefore the matrix $G^<_{n\lambda} = \langle n|(G\Sigma g)^<|\lambda\rangle$ can be recast as $G^<_{n\lambda} = \langle n|(G_C\ \Sigma_{CL}\ \tilde{g}_L)^<|\lambda\rangle$, or similarly with an explicit summation:

$$\begin{aligned} G^<_{n\lambda} =& \sum_{m,\lambda'} G^r_{nm}\ \Sigma^r_{m\lambda'}\ \tilde{g}^<_{\lambda'\lambda} \\ &+ G^r_{nm}\ \Sigma^{\mathrm{MB},<}_{m\lambda'}\ \tilde{g}^a_{\lambda'\lambda} + G^<_{nm}\ \Sigma^a_{m\lambda_2}\ \tilde{g}^a_{\lambda'\lambda}. \end{aligned} \tag{C7}$$

We also find that

$$\begin{aligned} G^<_{\lambda n} &= \langle\lambda|(\tilde{g}_L\ \Sigma_{LC}\ G_C)^<|n\rangle, \\ G^a_{n\lambda} &= \langle n|(G_C\ \Sigma_{CL}\ \tilde{g}_L)^a|\lambda\rangle, \\ G^r_{\lambda n} &= \langle\lambda|(\tilde{g}_L\ \Sigma_{LC}\ G_C)^r|n\rangle. \end{aligned} \tag{C8}$$

Using the rules of analytical continuation for products of three quantities, we find that Eq.(C4) becomes

$$\begin{aligned} &\sum_{\lambda n}\langle\lambda|\Sigma^r_{LC}|n\rangle\ \langle n|(G_C\ \Sigma_{CL}\ \tilde{g}_L)^<|\lambda\rangle - \langle\lambda|(\tilde{g}_L\ \Sigma_{LC}\ G_C)^<|n\rangle\ \langle n|\Sigma^a_{CL}|\lambda\rangle + \langle\lambda|\Sigma^{\mathrm{MB}<}_{LC}|n\rangle\ \langle n|(G_C\ \Sigma_{CL}\ \tilde{g}_L)^a|\lambda\rangle \\ &\quad - \langle\lambda|(\tilde{g}_L\ \Sigma_{LC}\ G_C)^r|n\rangle\ \langle n|\Sigma^{\mathrm{MB}<}_{CL}|\lambda\rangle \\ =&\sum_n\langle n|\left(G^<_C\ \Sigma^a_{CL}\ \tilde{g}^a_L + G^r_C\ \Sigma^<_{CL}\ \tilde{g}^a_L + G^r_C\ \Sigma^r_{CL}\ \tilde{g}^<_L\right)\Sigma^r_{LC}|n\rangle + \langle n|G^a_C\ \Sigma^a_{CL}\ \tilde{g}^a_L\ \Sigma^<_{LC} - \Sigma^<_{LC}\ \tilde{g}^r_L\ \Sigma^r_{LC}\ G^r_C|n\rangle \\ &\quad + \langle n|\Sigma^a_{CL}\left(\tilde{g}^<_L\ \Sigma^a_{LC}\ G^a_C + \tilde{g}^r_L\ \Sigma^<_{LC}\ G^a_C + \tilde{g}^r_L\ \Sigma^r_{LC}\ G^<_C\right)|n\rangle \\ =&\sum_n\langle n|G^r_C\left(\Sigma^<_{CL}\ \tilde{g}^a_L\ \Sigma^r_{LC} + \Sigma^r_{CL}\ \tilde{g}^<_L\ \Sigma^r_{LC} - \Sigma^<_{CL}\ \tilde{g}^r_L\ \Sigma^r_{LC}\right) + G^a_C\left(\Sigma^a_{CL}\ \tilde{g}^a_L\ \Sigma^<_{LC} - \Sigma^a_{CL}\ \tilde{g}^<_L\ \Sigma^a_{LC} - \Sigma^a_{CL}\ \tilde{g}^r_L\ \Sigma^<_{LC}\right) \\ &\quad + G^<_C\left(\Sigma^a_{CL}\ \tilde{g}^a_L\ \Sigma^r_{LC} - \Sigma^a_{LC}\ \tilde{g}^r_L\ \Sigma^r_{LC}\right)|n\rangle \\ =&\ \mathrm{Tr}_n\left[G^r_C\tilde{\Upsilon}^l_{LC} + G^a_C(\tilde{\Upsilon}^l_{LC})^\dagger + G^<_C(\tilde{\Upsilon}_{LC} - \tilde{\Upsilon}^\dagger_{LC})\right]. \end{aligned} \tag{C9}$$

In the second equality of Eq. (C9), the matrix elements have been swapped to get a trace only over the central region subspace $\{n\}$. The final two equalities are just exactly the Tr_n entering the definition of the current I_L given by Eq.(4) with the definitions of $\tilde{\Upsilon}^l_{LC}$ and $\tilde{\Upsilon}_{LC}$ (and their adjoints) given by

$$\begin{aligned}\tilde{\Upsilon}^l_{LC} &= \Sigma^<_{CL}\,(\tilde{g}^a_L - \tilde{g}^r_L)\,\Sigma^r_{LC} + \Sigma^r_{CL}\;\tilde{g}^<_L\;\Sigma^r_{LC} \\ &= (\Sigma\tilde{g})^<_{CL}\;\Sigma^r_{LC} - \Sigma^<_{CL}\;(\tilde{g}\Sigma)^r_{LC},\end{aligned} \tag{C10}$$

$$\tilde{\Upsilon}_{LC} = \Sigma^a_{CL}\;\tilde{g}^a_L\;\Sigma^r_{LC},$$

and

$$\begin{aligned}(\tilde{\Upsilon}^l_{LC})^\dagger &= \Sigma^a_{CL}\,(\tilde{g}^a_L - \tilde{g}^r_L)\,\Sigma^<_{LC} - \Sigma^a_{CL}\;\tilde{g}^<_L\;\Sigma^a_{LC} \\ &= (\Sigma\tilde{g})^a_{CL}\;\Sigma^<_{LC} - \Sigma^a_{CL}\;(\tilde{g}\Sigma)^<_{LC},\end{aligned} \tag{C11}$$

$$\tilde{\Upsilon}^\dagger_{LC} = \Sigma^a_{CL}\;\tilde{g}^r_L\;\Sigma^r_{LC}.$$

(QED) Eqs. (C3,C9,C10,C11) are just the main results of this paper.

Now we can follow the same mechanism of derivation to obtain an expression similar to Eq. (4) for the current I_R flowing at the right CR interface. In concrete the expression for I_R is given by Eq. (4) by swapping the index $L \leftrightarrow R$ and with a minus sign because of the current conservation condition $I_L + I_R = 0$.

Finally one should note that because of the following three conditions: *(i)* the very existence of the interaction crossing at the contact, *(ii)* the fact that in the most general cases $\Sigma^a_{C\alpha/\alpha C} \neq \Sigma^r_{C\alpha/\alpha C}$, in opposition with the non-interaction case where $V^a_{C\alpha/\alpha C} = V^r_{C\alpha/\alpha C} = V_{C\alpha/\alpha C}$, and *(iii)* the rules of analytical continuation for triple products $P_{(3)}$, the usual cyclic permutation performed in the calculation of the trace $\mathrm{Tr}_\lambda[(\Sigma G)^< - (G\Sigma)^<]$ cannot be used to transform the initial trace over $\{\lambda\}$ onto a trace over $\{n\}$. Therefore the current I_L at the LC contact is not given by a straightforward generalisation of the Meir and Wingreen formula of the type

$$\begin{aligned}I_L \neq \frac{e}{\hbar}\int\frac{d\omega}{2\pi}&\mathrm{Tr}_n[Y^<_L G^>_C - Y^>_L G^<_C] \\ &+ \mathrm{Tr}_\lambda[\Sigma^{\mathrm{MB}>}_L G^<_L - \Sigma^{\mathrm{MB}<}_L G^>_L].\end{aligned} \tag{C12}$$

where Y^x_L is the generalised (interacting) embedding potential of the L electrode.

This is another very important result of our work which has strong implication in the expression of the current itself, and also in the conditions of current conservation.

Appendix D: Current conservation condition

First we consider the general definition of the lesser and greater Green's functions :

$$G^{\gtrless} = (1 + G^r\Sigma^r)g^{\gtrless}(1 + \Sigma^a\;G^a) + G^r\;\Sigma^<\;G^a. \tag{D1}$$

The first term represents the initial conditions $g^{\gtrless}$.

For the central region, we have chosen the initial condition such as $\langle n|g^<|m\rangle = 0$ (see Eq.(10)). We could have chosen another initial condition. Such choices have no effects on the steady state regime when a steady current flow through the central region, however the initial conditions play an important role in the transient behaviour of the current[32–35].

For the definition of the lesser left and right Green's functions, it is however not possible to neglect the initial conditions (before full interactions and coupling to the region central are taken into account). This is because it would not be physical to ignore the presence of the left and right Fermi seas, obtained as the thermodynamical limit of the two semi-infinite leads which act as electron emitter and collector in our model device.

One can however recast Eq.(D1) as follows

$$G^< = G^r\;((g^r)^{-1}g^<(g^a)^{-1} + \Sigma^<)\;G^a = G^r\;\bar{\Sigma}^<\;G^a \tag{D2}$$

with $\bar{\Sigma}^< = \Sigma^< + \gamma^<$ and $\gamma^< = (g^r)^{-1}g^<(g^a)^{-1}$. And similarly for $G^>$. Hence $\gamma^< - \gamma^> = (g^a)^{-1} - (g^r)^{-1}$ and $\bar{\Sigma}^< - \bar{\Sigma}^> = (G^a)^{-1} - (G^r)^{-1}$.

From these properties, it can be easily shown that

$$\mathrm{Tr}_{\mathrm{all}}\left[\bar{\Sigma}^< G^> - \bar{\Sigma}^> G^<\right] = 0 \tag{D3}$$

for each ω. The trace runs over all indexes in the system (all $\equiv \{\lambda, n, \rho\}$) and the interaction Σ are spread over the whole L, C, R regions. This is the starting point to find the conditions for current conservation.

Because the trace runs over all the three subspaces, we can apply the usual cyclic permutation and recast Eq.(D3) as follows

$$-\mathrm{Tr}_{\mathrm{all}}\left[(\Sigma G)^< - (G\Sigma)^<\right] + \mathrm{Tr}_{\mathrm{all}}\left[\gamma^< G^> - \gamma^> G^<\right] = 0 \tag{D4}$$

or equivalently

$$\int \mathrm{d}\omega\;\mathrm{Tr}_{\mathrm{all}}\left[(\Sigma G)^< - (G\Sigma)^<\right] + \mathrm{Tr}_{\mathrm{all}}\left[\gamma^> G^< - \gamma^< G^>\right] = 0 \tag{D5}$$

Expanding the trace in the first term over each subspace $\mathrm{Tr}_{\mathrm{all}}[...] = \mathrm{Tr}_\lambda[..] + \mathrm{Tr}_n[...] + \mathrm{Tr}_\rho[...]$, one can identify the definition of the currents I_L and I_R from $\mathrm{Tr}_\lambda[...]$ and $\mathrm{Tr}_\rho[...]$ respectively.

Hence the condition of current conservation $I_L + I_R = 0$ leads to

$$\int \mathrm{d}\omega\;\mathrm{Tr}_{\mathrm{n}}\left[(\Sigma G)^< - (G\Sigma)^<\right] + \mathrm{Tr}_{\mathrm{all}}\left[\gamma^> G^< - \gamma^< G^>\right] = 0. \tag{D6}$$

After further manipulation, lengthy but trivial in the light of Appendix C, we find that the current conservation leads to the following condition:

$$\int \mathrm{d}\omega\;\mathrm{Tr}_{\alpha=L,C,R}[\Sigma^{\mathrm{MB}>}_\alpha G^<_\alpha - \Sigma^{\mathrm{MB}<}_\alpha G^>_\alpha] + C_{LC} + C_{CR} = 0 \tag{D7}$$

with

$$C_{LC}(\omega) = \mathrm{Tr}_n[(\tilde{Y}_L^{>} + \tilde{\Upsilon}_{LC} - \tilde{\Upsilon}_{LC}^{\dagger} - \tilde{\Upsilon}_{LC}^{l})G_C^{<} - (\tilde{Y}_L^{<} - \tilde{\Upsilon}_{LC}^{l})G_C^{>} + (\tilde{\Upsilon}_{LC}^{l} + (\tilde{\Upsilon}_{LC}^{l})^{\dagger})G^{a}] \quad \text{(D8)}$$

and $C_{CR} = C_{LC}[\{L \leftrightarrow R\}]$.

The first trace in Eq. (D7) corresponds to the sum over the three regions L, C, R of the integrated collision term. The two other traces C_{LC} and C_{CR} arise from the interactions crossing at the LC and CR interfaces. But, globally, Eq. (D7) still implies that the total integrated collision terms must vanish.

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